Pairing in Cu-O Models: Clues of Joint Electron-Phonon and Electron-Electron Interactions

Enrico Perfetto and Michele Cini
Istituto Nazionale di Fisica Nucleare, Dipartimento di Fisica, Universita' di Roma Tor Vergata, Via della Ricerca Scientifica, 1-00133 Roma, Italy

We discuss a many-electron Hamiltonian with Hubbard-like repulsive interaction and linear coupling to the phonon branches, having the Cu-O plane of the superconducting cuprates as a paradigm. A canonical transformation extracts an effective two-body problem from the many-body theory. As a prototype system we study the CuO_4 cluster, which yields electronic pairing in the Hubbard model; moreover, a standard treatment of the Jahn-Teller effect predicts distortions that destroy electronic pairing. Remarkably, calculations that keep all the electronic spectrum into account show that vibrations are likely to be synergic with electronic pairing, if the coupling to half-breathing modes predominates, as experiments suggest.

73.22.-f Electronic structure of nanoscale materials: clusters, nanoparticles, nanotubes, and nanocrystals
74.20.Mn Nonconventional mechanisms
71.27.+a Strongly correlated electron systems; heavy fermions

The role of electron-phonon (EP) interactions in determining the superconducting correlations in the Cu-O planes of cuprates is a very controversial issue. Possibly, the pairing mechanism has a predominantly electronic origin [1], but many high-T_C compounds exhibit a quite noticeable doping dependent isotope effect [2], suggesting that EP interactions could be important and should be included in the theory. In particular there is experimental evidence [3] that the half breathing Cu-O bond stretching mode at $k = (\pi, 0), (0, \pi)$ is significantly coupled with the doped holes in the superconducting regime and its contribution may be relevant for the $d_{x^2-y^2}$ pairing [4] [5] [6].

The simplest way to include both strong electronic correlations and EP interactions is the Hubbard-Holstein model, where electrons are coupled to a local Einstein phonon. Much is known [7] [8] about the possibility of a superconducting phase in this model; further evidence for pairing can be extracted by exact numerical diagonalization of small cluster Hamiltonians, calculating the effective pairing interaction defined as $\tilde{\Delta}(N+2) = E(N+2) + E(N) - 2E(N+1)$, where N is the number of fermions in the system. A negative $\tilde{\Delta}$ is interpreted as an effective attraction and suggests the presence af a bound pair in the ground state. Petrov and Egami [9] found $\tilde{\Delta} < 0$ in a doped 8-site Hubbard-Holstein ring and showed that the effect disappears once phonons are turned off. Conversely Mazumdar and coworkers [10] suggested that pairing in Hubbard clusters reported by some authors [11] was of doubtful physical interpretation due to the neglect of the lattice degrees of freedom and the Jahn-Teller (JT) effect. Consider even N, such that the $N+2$-particle cluster hosts a singlet pair and the $N+1$-particle ground state is degenerate. They argued that JT distorsion might cause a larger energy gain of the system with $N+1$ particles, thus reversing the sign of $\tilde{\Delta}$. This led to the conjecture that any $\tilde{\Delta} < 0$ due to an electronic mechanism is just a finite size effect, which vanishes for large systems like the JT effect does.

Here we further investigate this issue by addressing the question if the recently proposed $W = 0$ pairing avaliable in the Hubbard model [12] [13] [14] survives when the lattice degrees of freedom are switched on. In previous works it was shown that a class of C_{4v}-symmetric clusters exhibit the $\tilde{\Delta} < 0$ property, due to the binding of the so called $W = 0$ pairs. This depends crucially on the symmetry and on the availability of degenerate states. Fermions in strongly correlated systems can avoid the on-site repulsion and possibly form singlet bound states if there is enough symmetry; the fillings and the symmetry channels where the $W = 0$ pairing can occur are determined in full generality by the $W = 0$ theorem [15]; these symmetries achieve the same result as high angular momentum and parallel spins in the Kohn-Luttinger [16] continuum approach.

When lattice effects are introduced in this scenario, several questions arise. In the conventional mechanism, phonons overscreen the electron repulsion; what happens if electronic screening already leads to pairing? It is not obvious that the phonons will reinforce the attraction while preserving the symmetry. More generally, some vibrations could be pairing and others pair-breaking. To address these problems we use an extension of the Hubbard model in which bond stretchings dictate the couplings to the normal modes of the C_{4v}-symmetric configuration, generating a long-range (Fröhlich) EP interaction. This is physically more detailed than the Hubbard-Holstein model, and does not restrict to on-site EP coupligs that would be impaired by a strong Hubbard repulsion. A further important drawback of the Holstein EP interaction in this context is an unphysically large polaron (and bipolaron) mass and low polaron mobility. This problem is avoided using Fröhlich-like phonons for modeling the Cu-O planes. It was pointed out recently [17] [18] [19] that even in the strong EP coupling regime the a long-range EP interaction removes the problem of polaron self-trapping.

We start from the Hubbard model with on-site interaction U and expand the hopping integrals $t_{i,j}(\mathbf{r}_i, \mathbf{r}_j)$ in powers of the displacements ρ_i around a C_{4v}-symmetric equilibrium configuration

$$t_{i,j}(\mathbf{r}_i,\mathbf{r}_j) \simeq t^0_{i,j}(\mathbf{r}_i,\mathbf{r}_j) + \sum_\alpha \left[\frac{\partial t_{ij}(\mathbf{r}_i,\mathbf{r}_j)}{\partial r_i^\alpha}\right]_0 \rho_i^\alpha + + \sum_\alpha \left[\frac{\partial t_{ij}(\mathbf{r}_i,\mathbf{r}_j)}{\partial r_j^\alpha}\right]_0 \rho_j^\alpha \,, \quad (1)$$

where $\alpha = x, y$. Below, we write down the ρ_i^α in terms of the normal modes $q_{\eta\,\nu}$: $\rho_i^\alpha = \sum_{\eta\,\nu} S^\alpha_{\eta\,\nu}(i)\, q_{\eta\,\nu}$, where η is the label of an irreducible representation (*irrep*) of the symmetry group of the undistorted system and ν is a phonon branch.

Thus, treating the Cu atoms as fixed, for simplicity, one can justify an electron-lattice Hamiltonian:

$$H_{el-latt} = H_0 + V_{\rm tot}\,. \quad (2)$$

Here $H_0 = H_0^n + H_0^e$ is given by

$$H_0 = \sum_\eta \hbar\omega_{\eta,\nu} b^\dagger_{\eta,\nu} b_{\eta,\nu} + \sum_{i,j\sigma} t^0_{i,j}(\mathbf{r}_i,\mathbf{r}_j)(c^\dagger_{i\sigma} c_{j\sigma} + h.c), \quad (3)$$

where $\omega_{\eta,\nu}$ are normal mode frequencies. Moreover, let M denote the O mass, $\xi_{\eta,\nu} = \lambda_{\eta\,\nu}\sqrt{\frac{\hbar}{2M\omega_{\eta,\nu}}}$, with $\lambda_{\eta\,\nu}$ numbers of order unity that modulate the EP coupling strength. Then, $V_{\rm tot} = V + W$ reads

$$V_{\rm tot} = \sum_{\eta,\nu} \xi_{\eta,\nu}(b^\dagger_{\eta,\nu} + b_{\eta,\nu}) H_{\eta,\nu} + U\sum_i n_{i\uparrow} n_{i\downarrow}, \quad (4)$$

the $H_{\eta,\nu}$ operators are given by

$$H_{\eta,\nu} = \sum_{i,j}\sum_{\alpha,\sigma} \left\{ S^\alpha_{\eta\,\nu}(i)\left[\frac{\partial t_{ij}(\mathbf{r}_i,\mathbf{r}_j)}{\partial r_i^\alpha}\right]_0 + S^\alpha_{\eta\,\nu}(j)\left[\frac{\partial t_{ij}(\mathbf{r}_i,\mathbf{r}_j)}{\partial r_j^\alpha}\right]_0 \right\} (c^\dagger_{i,\sigma} c_{j,\sigma} + h.c.)\,. \quad (5)$$

Consider the Hubbard model $H_H = H_0^e + W$ involving the purely electronic terms of $H_{el-latt}$. We recall that $W = 0$ pairs are defined as two-body singlet eigenstates of the H_H free of double occupancy; in the many-body problem they interact indirectly and can get bound [12] [20]. Here we wish to derive an effective interaction between the particles in the pair suitable for $H_{el-latt}$, by generalizing the canonical transformation approach of Ref. [20].

Denote the phonon vacuum by $|0\rangle\rangle$ and the N-particle non-interacting Fermi sphere by $|\Phi_0(N)\rangle$; if we add a $W = 0$ pair to $|\Phi_0(N)\rangle \otimes |0\rangle\rangle$, the two extra particles, by definition, cannot interact directly (in first-order). Hence their effective interaction comes out from virtual electron-hole (e-h) excitation and/or phonon exchange and in principle can be attractive. To expand the interacting $(N+2)$-fermions ground state $|\Psi_0(N+2)\rangle$, we build a complete set $\mathcal{S}$ of configurations in the subspace with vanishing z spin component, considering the vacuum state $|\Phi_0(N)\rangle \otimes |0\rangle\rangle$ and the set of excitations over it.

We start by creating $W = 0$ pairs of fermions over $|\Phi_0(N)\rangle\otimes|0\rangle\rangle$; we denote with $|m\rangle\otimes|0\rangle\rangle$ these states. At weak coupling, we may truncate the Hilbert space to the simplest excitations, i.e., to states involving 1 e-h pair or 1 phonon created over the $|m\rangle\otimes|0\rangle\rangle$ states. We define the $|m\rangle\otimes|q\rangle\rangle$ states, obtained by creating a phonon denoted by $q = (\eta,\nu)$ over the $|m\rangle \otimes |0\rangle\rangle$ states. Finally we introduce the $|\alpha\rangle \otimes |0\rangle\rangle$ states, obtained from the $|m\rangle\otimes|0\rangle\rangle$ states by creating 1 electron-hole (e-h) pair. We now expand the interacting ground state in the truncated Hilbert space:

$$|\Psi_0(N+2)\rangle = \sum_m a_m |m\rangle\otimes|0\rangle\rangle + + \sum_{m,q} a_{m,q}|m\rangle\otimes|q\rangle\rangle + \sum_\alpha a_\alpha |\alpha\rangle\otimes|0\rangle\rangle \quad (6)$$

and set up the Schrödinger equation with energy eigenvalue E. A canonical transformation [21] decouples the higher amplitudes $a_{m,q}$ and a_α, and the a_m become expansion coefficients over the $W = 0$ pairs of the wave function of the dressed pair $|\varphi\rangle$. This obeys the Cooper-like equation $H_{\rm pair}|\varphi\rangle = E|\varphi\rangle$ with an effective two-body Hamiltonian $H_{\rm pair}$:

$$H_{\rm pair} \equiv (H_0 + W + S[E]) \quad (7)$$

where S is the $E-$dependent effective scattering operator; the pairing problem must be solved self-consistently. It turns out that the effective scattering operator is

$$S[E]_{m,m'} = \sum_\alpha \frac{W_{m,\alpha}W_{\alpha,m'}}{E'_\alpha - E} + \sum_{m'',q} \frac{V^q_{m,m''}V^q_{m'',m'}}{E'_{m''} + \omega'_q - E}\,. \quad (8)$$

Here the primed quantites in the denominators are the eigenenergies of the $|m\rangle\otimes|0\rangle\rangle$, $|m\rangle\otimes|q\rangle\rangle$, $|\alpha\rangle\otimes|0\rangle\rangle$ states; they get renormalized in the decoupling procedure. The pairing criterion involves the properly renormalized Fermi energy ε_F^R (see ref. [13]); if the lowest energy eigenvalue E is such that $E = 2\varepsilon_F^R + \Delta$ with negative Δ, the dressed $W = 0$ pair gets bound in the many-body interacting problem and the system undergoes a Cooper instability.

As an illustrative application of the above pairing scheme, in this preliminary work we focus on CuO_4, the smallest cluster yielding $W = 0$ pairing in the Hubbard model. This requires 4 holes, (total number, not referred to half filling); such a doping is somewhat unrealistic, but larger C_{4v}-symmetric clusters and the full CuO_2 plane also show $W = 0$ pairing in the doping regime relevant for cuprates [12] [20]. Remarkably, in the pure Hubbard model, one can verify that $\Delta = \tilde{\Delta}(4)$ at least at weak coupling [12], which demonstrates that $\tilde{\Delta}$ has the physical meaning of an effective interaction. CuO_4 represents a good test of the interplay between electronic and phononic pairing mechanisms since we can compare exact diagonalization results with the analytic approximations of the canonical transformation. CuO_4 allows only the coupling to phonons at the centre or at the edge of the Brillouin Zone; however, phonons near the edge are precisely those most involved [3] [4].

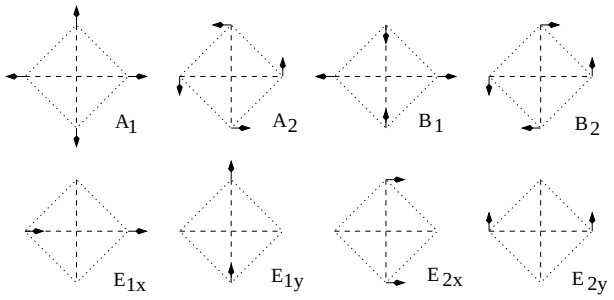

FIG. 1. Pictorial representation of the ionic displacements in the eight normal modes of the CuO4 cluster, labelled according to the *irreps* of the C_{4v} Group.

Even in this small system the virtually exact diagonalizations are already hard and the next C_{4v}-symmetric example, the Cu_5O_4 cluster [22], is much more demanding for the number of vibrations and the size of the electronic Hilbert space. The pure Hubbard CuO_4 cluster with O-O hopping $t_{ox} = 0$ yields [12] $\tilde{\Delta}(4) < 0$, due to a couple of degenerate $W = 0$ bound pairs, in the A_1 and B_2 *irreps* of the C_{4v} group; therefore in Eq.(8) we set the $m = m'$ labels accordingly. Starting with the C_{4v}-symmetric arrangement, any displacement of the Oxygens in the plane can be analised in *irreps*, $A_1, A_2, B_1, B_2, E_1, E_2$, see Fig.1. The EP coupling is expressed through two parameters ∂t and ∂t_{ox}, defined as the derivatives of t and t_{ox} with respect to the Cu-O and O-O bond lengths. The CuO_4 cluster is also interesting as a test of the conjecture of Ref. [10]; we checked [21] that a standard JT calculation in which the degenerate ground state wave functions interact with the vibrations indeed predicts distortions that already at moderate EP coupling destroy $W = 0$ pairing; here we wish to go beyond this approximation.

According to Eq.(8), the phonon-mediated interaction, in the A_1 channel is:

$$\sum_{m'',q} \frac{V^q_{A_1,m''} V^q_{m'',A_1}}{E_{m''} + \omega'_q - E} = -\frac{4}{3}\partial t^2_{ox} \left(\frac{\lambda^2_{B_2}}{2\varepsilon_{A_1} + \omega_{B_2} - E} + \frac{2\lambda^2_{A_1}}{2\varepsilon_{A_1} + \omega_{A_1} - E} - \frac{\lambda^2_{E_1}}{2\varepsilon_{A_1} + \omega_{E_1} - E} - \frac{\lambda^2_{E_2}}{2\varepsilon_{A_1} + \omega_{E_2} - E} \right). \quad (9)$$

In the B_2 channels we find:

$$\sum_{m'',q} \frac{V^q_{B_2,m''} V^q_{m'',B_2}}{E'_{m''} + \omega'_q - E} = -4\partial t^2_{ox} \frac{\lambda^2_{B_2}}{2\varepsilon_{A_1} + \omega_{B_2} - E} \,. \quad (10)$$

Thus, the two $W = 0$ pairs behave differently: the B_2 binding energy is enhanced by phonons, while in the A_1 sector the overall sign depends on λ_η and ω_η. It turns out that A_1 and B_2 modes are synergic to the $W = 0$ pairing, while both longitudinal and transverse E modes are pair-breaking. The half-breathing modes that are deemed most important [4] [5] are $A_1 \pm B_1$ combinations, but B_1 is scarcely relevant. For the sake of argument, in the explicit calculations we took all the normal modes with the same energy $\varepsilon_0 = \hbar\omega_0 = 10^{-1}$eV and $\lambda_\eta = 1$. This sets the length scale of lattice effects $\xi_0 = \sqrt{\frac{\hbar}{2M\omega_0}} \simeq 10^{-1}$ Å where we used $M = 2.7 \times 10^{-26}$Kg for Oxygens.

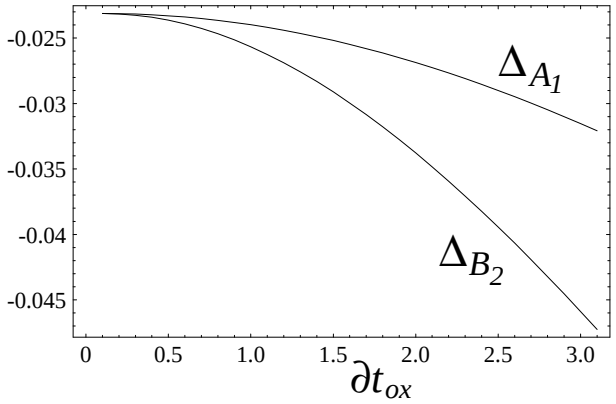

FIG. 2. Analytical results of the canonical transformation: pair binding energy in the A_1 and B_2 sectors as a function of ∂t_{ox}. Here we used $\lambda_\eta = 1$ for every mode, $t = 1$eV, $t_{ox} = 0$, $U = 1$eV; ∂t_{ox} is in units of $\varepsilon_0/\xi_0 = 1\text{eV} \times$ Å^{-1}, Δ is in eV.

With this choice, pairing is enhanced in the A_1 sector as well, albeit less than in B_2, see Fig.2, where the pairing energy without phonons is $\Delta \simeq -20$meV for both the $W = 0$ pairs. In other terms, the vibrations split the degeneracy of the electronic ground state, effectively lowering the symmetry like a nonvanishing t_{ox}. Here again we stress that $\Delta = \tilde{\Delta}(4)$ for both symmetries, generalizing the Hubbard model result. To go beyond the weak coupling case, we diagonalized $H_{el-latt}$ numerically in the truncated Hilbert space with up to 5 modes at a time having vibrational quantum numbers $n_v \leq 3$. In Figure 3a we included all vibrations except E_1; with increasing the EP coupling, the pair binding energy steadily increases, in agreement with the trend in Fig.2; the unbinding mode E_2 is dominated by the pairing ones. Note that the binding energy increases with ∂t_{ox} as predicted by the weak coupling theory, but also with $|\partial t|$, suggesting that at strong coupling the Cu-O bond stretching grows in importance.

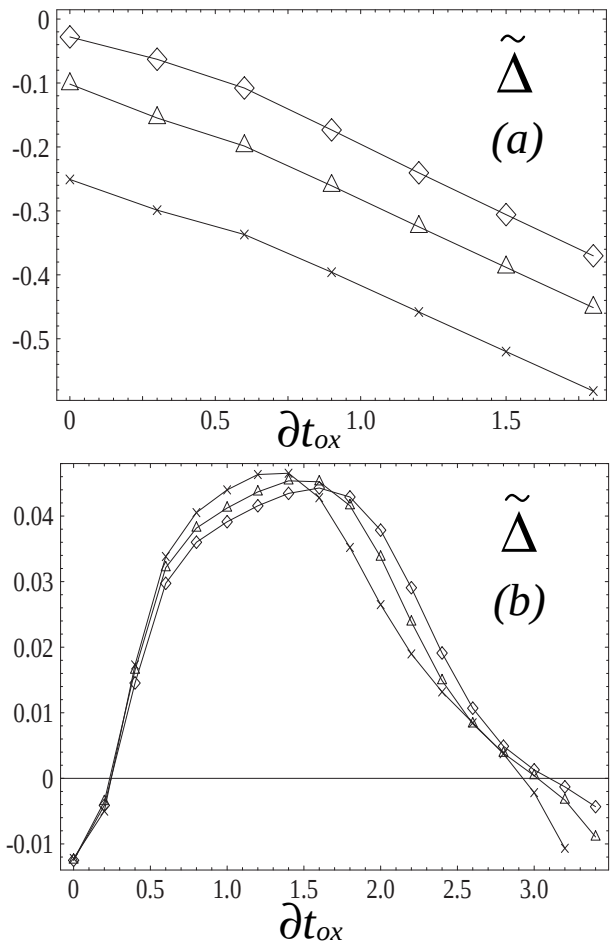

FIG. 3. $\tilde{\Delta}(4)$ as a function of ∂t_{ox} for different values of ∂t. $\lambda_\eta = 1$ all the vibrations, except: $\lambda_\eta = 0$ for $\eta = E_1$ (a); $\lambda_\eta = 0$ for $\eta = E_2$ (b). n_v=3 for each active mode; $\partial t = -0.2$ (diamonds); $\partial t = -0.5$ (triangles); $\partial t = -1$ (crosses). Here we used $t = 1$eV, $t_{ox} = 0$, $U = 1$eV, ∂t_{ox} and ∂t are in units of $\varepsilon_0/\xi_0 = 1\text{eV}\times \text{Å}^{-1}$, $\tilde{\Delta}$ is in eV.

In Figure 3b we included all vibrations except E_2. The E_1 vibrations are strongly pair breaking, and $\tilde{\Delta}(4)$ is much smaller in magnitude than in Figure 3a; it becomes positive at intermediate coupling. It is likely that the couplings to the pair-breaking E modes are somewhat underestimated by the choice of λ parameters in Fig.3a and overestimated in Fig.3b. Interestingly, however, at stronger coupling the pairing modes prevail again driving the system to pairing (bipolaron formation). This resembles the trend found in Ref [9], although there a Hubbard-Holstein framework is used and since the system is one-dimensional the electronic pairing does not occur.

These results yield several indications. First, the catastrophic distortions predicted by the JT approximation are not borne out by more complete approaches involving a broader spectrum of electronic states. This interesting result arises since the electronic ground state belongs to a narrow multiplet; the standard treatment of the JT effect singles out the lowest level, which is justified only at weak EP coupling. Pairing prevails also at strong coupling in part of the parameter space, in the symmetry channels where $W = 0$ pairs occur. The correct trend is predicted by the canonical transformation approach, which also explains the pairing or pair-breaking character of the modes. In particular it is found that the half-breathing modes give a synergic contribution to the purely electronic pairing; since they are believed to be mainly involved in optimally doped cuprates, our findings suggest a joint mechanism, with the Hubbard model that captures a crucial part of the physics. This scenario was also drawn in the context of an extended t-J model, where the half-breathing mode was found to enhance electronic pairing [6] [23]. Finally, experimental data on nanopowders [24] also indicate that one should not be overly pessimistic about cluster calculations. $YBa_2Cu_3O_{6+x}$ powders consisting of 0.7 nm thick grains with a 11 nm radius show the same T_C as the bulk, indicating that going to the thermodynamic limit could not be essential to understand pairing. Cluster calculations can be relevant and physically insightful concerning the interplay of various degrees of freedom on pair structure and formation.

[1] For an overview, see E. Dagotto, *Rev. Mod. Phys.* **66**,763 (1994) and references therein.

[2] M. K. Crowford,M.N. Kunchur,W.E. Farneth,E.M. McCarron,S.J. Poon, *Phys. Rev.* **B 41**, 282 (1990); *Science* **250**, 1390 (1990); J.P. Franck et al.,*Physica C* **185-189**, 1379 (1991); *Phys. Rev. Lett.* **71**, 283 (1993); J.P. Franck, *Physica C* **235-240**,1503 (1994);

[3] R. J. McQueeney, Y. Petrov, T. Egami, M. Yethiraj, G. Shirane and Y. Endoh, *Phys. Rev. Lett.* **82**, 628 (1999).

[4] Z. X. Shen, A. Lanzara, S. Ishihara, and N. Nagaosa, cond-mat/0108381.

[5] O. Rösh and O. Gunnarsson, cond-mat/0308035.

[6] S. Ishihara and N. Nagaosa, cond-mat/0311200.

[7] C.H. Pao and H.B. Schuttler, *Phys. Rev.* **B 57**, 5051 (1998); *Phys. Rev.* **B 60**, 1283 (1999) .

[8] A.S. Alexandrov, *Phys. Rev.* **B 46**, 2838 (1992); G. Wellein, H. Roder and H. Fehske, *Phys. Rev.* **B 53**, 9666 (1996); J. Bonca, T. Katrasnik and S.A. Trugman, *Phys. Rev. Lett.* **84**, 3153 (2000).

[9] Y. Petrov and T. Egami, *APS, Annual March Meeting* **Y 20**, 003 (2000).

[10] S. Mazumdar, F. Guo, D. Guo, K. C. Ung and J. T. Gammel, *Proceeding of the "Discussion meeting on strongly correlated electron systems in chemistry*, Bangalore, India, (Springer Verlag 1996).

[11] S.R. White, S. Chakravarty, M.P. Gelfand and S.A. Kivelson , *Phys. Rev.* **B** 45, 5062 (1992); A. Parola, S. Sorella, M. Parrinello, and E. Tosatti *Phys. Rev.* **B** 43, 6190 (1991)

[12] M. Cini and A. Balzarotti, *Phys. Rev.* **B 56**, 14711 (1997).

[13] E. Perfetto, G. Stefanucci and M. Cini, *Phys. Rev.* **B 66**, 165434 (2002).

[14] Agnese Callegari, Michele Cini, Enrico Perfetto and Gianluca Stefanucci, *Phys. Rev.* B **68**, 153103, (2003).

[15] Michele Cini, Adalberto Balzarotti, Raffaella Brunetti, Maria Gimelli and Gianluca Stefanucci, *Int. J. Mod. Phys.* **B 14**, 2994 (2000).
[16] W. Kohn and J. M. Luttinger, *Phys. Rev. Lett.* **15**,524 (1965).
[17] J. Bonca and S. A. Trugman, *Phys. Rev.***B 64**, 094507 (2001).
[18] A. S. Alexandrov and P. E. Kornilovitch, *J. Phys.* **C 14**,5337 (2002).
[19] A. S. Alexandrov, *Int. J. Mod. Phys.* **B 17**, 3315 (2003).
[20] Michele Cini, Gianluca Stefanucci and Adalberto Balzarotti, *Eur. Phys. J.* **B10**, 293 (1999).
[21] E. Perfetto and M. Cini unpublished.
[22] Michele Cini, Adalberto Balzarotti and Gianluca Stefanucci, *Eur. Phys. J.* **B14**, 269 (2000).
[23] T. Sakai, D. Poilblanc, and D. J. Scalapino, *Phys. Rev.* **B 55**, 8445 (1997).
[24] P.Paturi, J.Raittila,H. Huhtinen, V-P Huhtala and R. Laiho, *J. Phys. Condensed Matter* **B 15** (2003) 2103.